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RAINOUT STUDIES AT LAWRENCE LIVERMORE LABORATORY

Joseph B. Knox
Allen L. Williams

February 11, 1974

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Abstract

The study of rainout has received additional impetus from recent investigations of the impact of collateral damage upon tactical nuclear operations. Additional research is going forward at Lawrence Livermore Laboratory to provide an improved technical basis for the assessment of rainout. The project is designed to develop improved understanding of the basic physical interactions that control the processes and to assess more rigorously the potential hazards to man. Aspects of the work described in this report include a microphysical description of precipitation scavenging of the nuclear debris aerosol, progress in the numerical modeling of natural cloud systems and their interactions with nuclear debris aerosols, and investigations of possible means of controlling the rainout-removal process. This is an interim report; it is expected that continuing research over the next six months will permit a more precise basis for analysis of the phenomenon.

Introduction

In a tactical nuclear conflict, the delayed radiation effects of close-in fallout would present complications from the viewpoint of collateral damage. Significant dry fallout can be avoided by exclusive use of free-air bursts, but wet deposition from precipitation scavenging of nuclear debris clouds is predictable, if at all, only shortly before the burst. The extent and location of the rainout radiation field would depend upon the timing and nature of the interaction of the debris cloud with the natural precipitation-scavenging environment. It turns out that rainout is a particular concern with the low-yield weapons characteristic of the tactical nuclear stockpile; high-yield weapons can carry nuclear debris above the altitudes conducive to precipitation scavenging.

The potential exposure to man from low-yield free-air bursts was assessed in UCRL-51164.[1] The major conclusion was that the hazard of close-in exposure from these bursts could be appreciable if scavenging by precipitation occurred. If the entire vertical integral of radioactivity contained in a nuclear debris cloud were brought down, then the potential infinite whole-body exposure vs distance from ground zero would be that approximated in Fig. 1. For example,

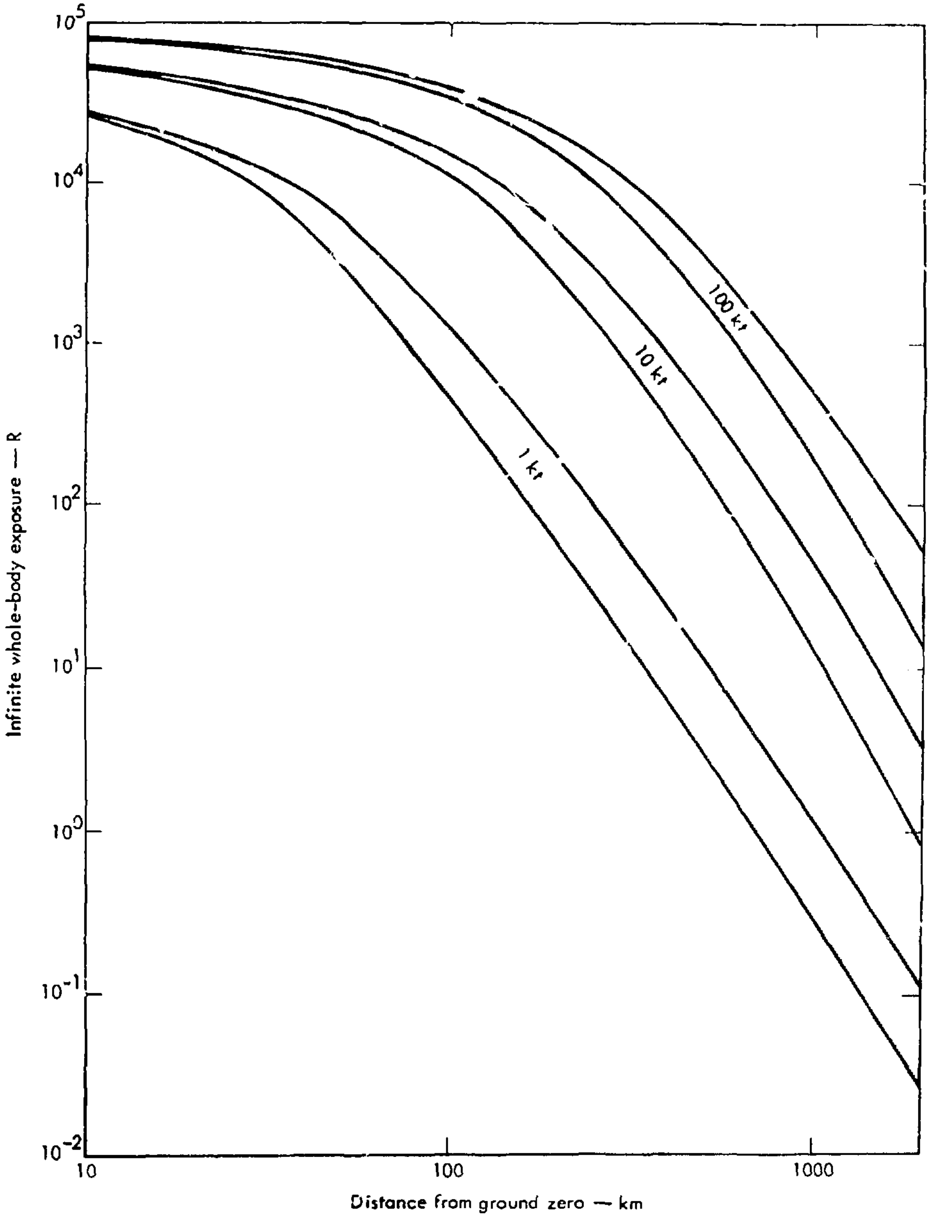

Fig. 1. Vertical integral (infinite whole-body exposure) due to gross gamma radiation as a function of distance from ground zero. The upper curve for each yield represents the case of slow horizontal diffusion; the lower curve represents the case of fast diffusion.

rainout of the radioactivity within the debris cloud of a 1-kt air burst could result at 100 km downwind in infinite whole-body doses of 1000 R and higher. Further work, published in UCRL-51328 (1972),[2] substantiated this previous assessment; however, major uncertainties remain in understanding the physical bases for rainout processes as well as in determining the frequency of occurrence of the conditions that would lead to radiation hazard by the rainout process.

Rainout is not a newly discovered phenomenon, but its study has received additional impetus from recent investigations of the impact of collateral damage upon tactical nuclear operations. Additional research is going forward at this Laboratory to provide an improved technical basis for the rainout assessment. This work, funded by the Defense Nuclear Agency and supplemented in part by the Laboratory's Whitney Program, is summarized herein. The project is designed to develop improved understanding of the basic physical interactions controlling the processes and to assess more rigorously the potential hazards to man.

Microphysical Description of Precipitation Scavenging

Aerosol particles are removed from the atmosphere by different processes depending on their size. Particles larger than about 1 μm can be scavenged by direct impaction with raindrops. This inertial capture process is the primary removal mechanism for washout or below-cloud scavenging, the washout efficiency being a function of particle size.

For rainout or in-cloud scavenging, other processes must be considered in addition to inertial capture. Aerosol particles with perfectly wettable surfaces and radii greater than 0.1 μm can serve as nucleation sites for the formation of cloud droplets. Soluble particles with radii as small as 0.01 μm can be nucleated in clouds. According to arguments by Fletcher[3] and experimental results by Twomey,[4] an insoluble particle whose surface departs from complete wettability, i.e., for which the contact angle is greater than say 6 deg, cannot serve as a condensation nucleus at typical cloud supersaturations. Nucleated particles can be deposited on the ground either through growth to raindrop size and falling (a very rare event) or by growing to several micrometers in radius and being accreted by a raindrop.

Particles of radius less than about 0.02 μm display considerable Brownian motion under atmospheric conditions, allowing them to collide and attach to cloud droplets or raindrops. This scavenging mechanism is directly effective only for short times after the particles are formed, since, given time, they will readily attach to larger debris particles or to natural aerosol particles. Those particles attached to droplets due to Brownian capture can be removed to the ground through accretion of the droplet.

Debris particles from free-air bursts consist mainly of oxides of the principal casing materials. The various radionuclides make up a small percentage of the total particle mass. Indications are that debris particles are insoluble in water and are wettable due to their large surface energy compared to that of water. Some of the radionuclides are soluble, but it is not clear whether the amount of soluble material on the particle surface can significantly change the nucleation characteristics as suggested by Hicks.[5]

Indications are that the particle sizes resulting from free-air bursts are distributed lognormally and that the mean particle size decreases with increasing yield.[6,7] It appears reasonable to expect low-yield air bursts to produce particles susceptible to inertial capture and/or nucleation scavenging. However, higher-yield free-air bursts may produce particles below the nucleation threshold, leaving Brownian scavenging as the dominant removal mechanism.

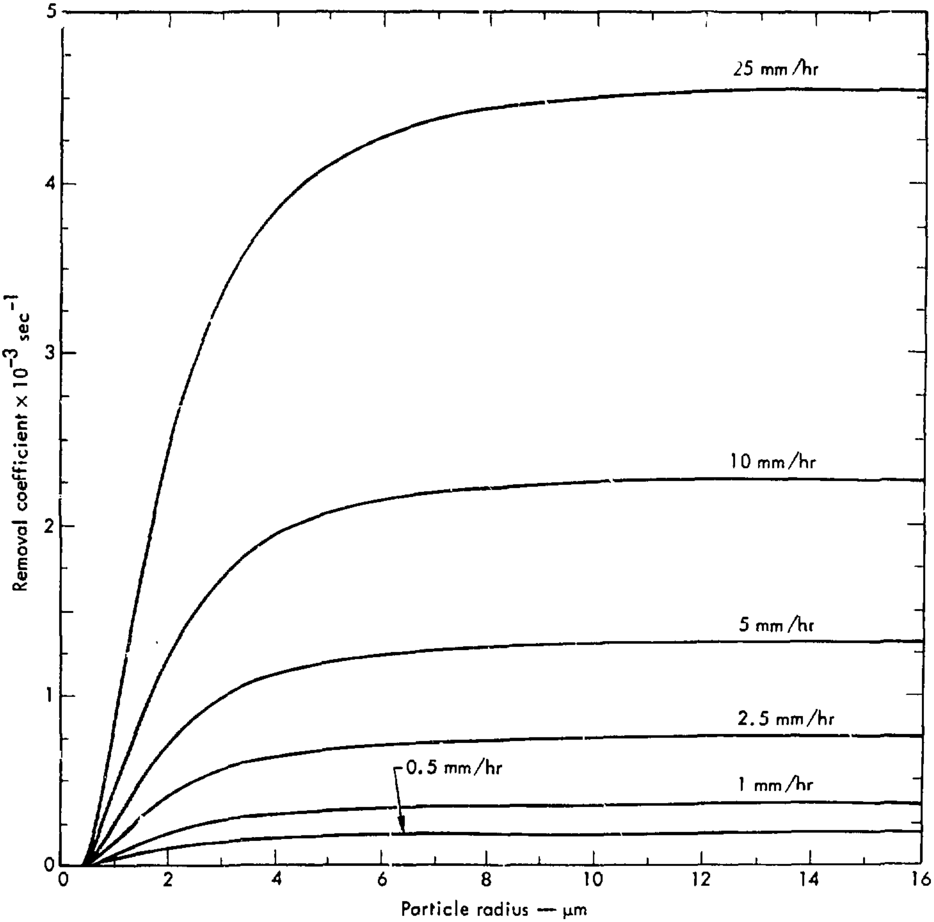

Fig. 2. Removal coefficient versus particle radius for various rain rates.

If we define $n(r, t)$ as the number of debris particles of radius r per unit volume of air at time t, the rate of removal of the particles by rain can be written as

$$\frac{dn(r, t)}{dt} = - \Lambda n(r, t). \qquad (1)$$

Here Λ represents the removal rate of one particle of radius r. Assuming that inertial capture is the removal mechanism, Fig. 2 gives Λ as a function of r for various rain rates. Note that Λ decreases rapidly as the particle radius falls below about 1 μm.

If the debris particle can act as a site for the droplet formation, it will grow by diffusion of water vapor on the surface to a radius of about 10 μm in a short period of time. The removal rate would then be given by that for a particle radius of 10 μm on Fig. 2. To gain an idea of the efficiency of these removal rates, assume that the part particles can act as efficient condensation nuclei. Then, using for Λ in Eq. 1 the removal rate corresponding to $r = 10$ μm in Fig. 2, we can obtain the fraction of particles removed from a volume of air as a function of time. The results of such a calculation are given in Fig. 3.

PARTICLE SIZE DISTRIBUTION

To improve our estimate of the removal rate of debris particles, it is important to know the particle size distribution. We have some experimental data on debris-particle size distributions from past U.S. atmospheric tests, but there are few data pertaining to low-yield devices. We are now developing a computational capability to calculate the particle size distribution of particles from low-yield free-air bursts. We use fairly well known expressions for the temperature and the volume as functions of time to determine the number of particles formed by condensation and then follow the particle growth to determine the final size distribution. Early results look encouraging. If the code can be validated against existing experimental data, taking into account the event detonation conditions, we will have a tool to predict particle size distributions for stockpile devices or devices in the design stage.

EXPERIMENT

We are planning to carry out a set of experiments this next summer. At the moment we plan to use two air platforms, manned and unmanned. The properties that we plan to measure are:

1. Particle size distribution and distribution of radionuclides as a function of particle size. As discussed above, knowledge of particle size distribution is necessary to determine the mechanism and the subsequent rate of removal by precipitation. Since the removal rates are dependent on particle size, one must know the amount of radioactivity as a function of particle size to determine the potential radiation hazard.

2. Nucleation characteristics. Debris particles are composed predominantly of oxides of the casing material, and when first formed they are highly wettable. The clean surfaces probably are contaminated when exposed to the atmosphere and as a

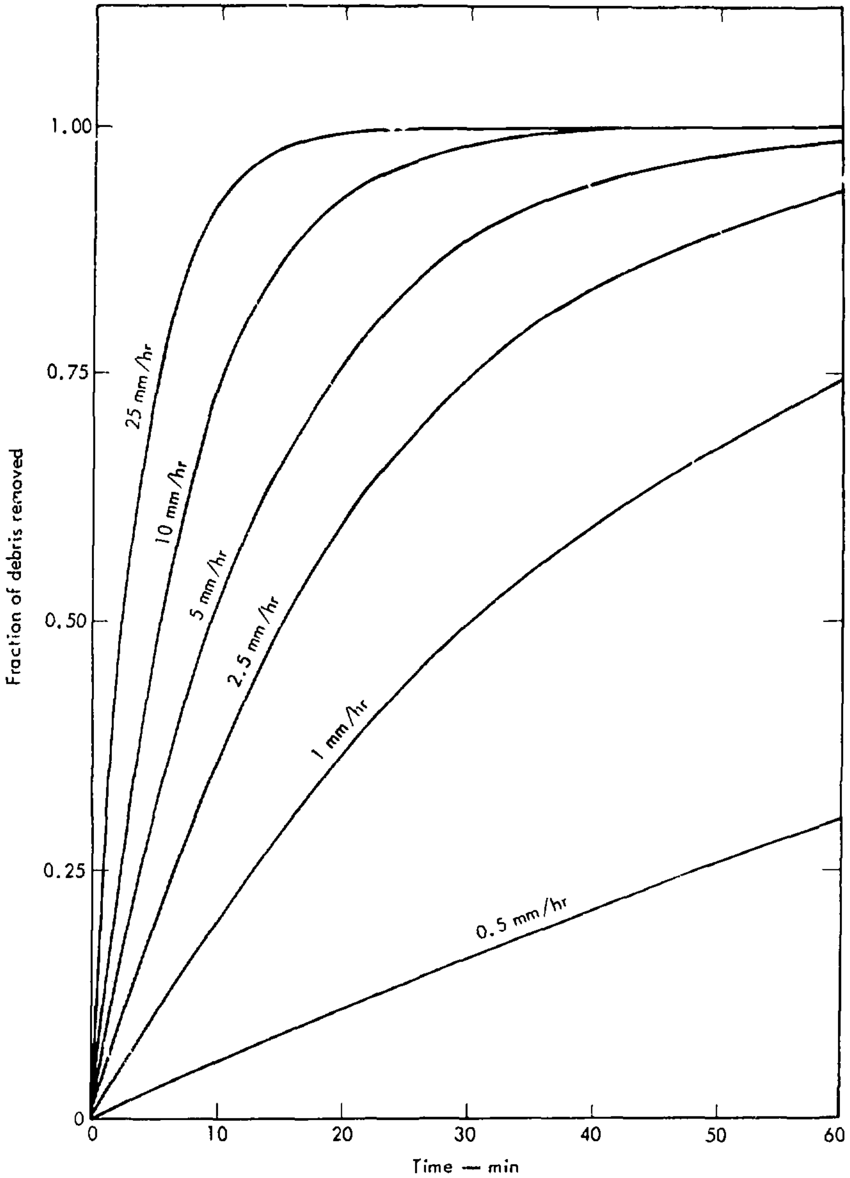

Fig. 3. Fraction of radioactivity originally in the rain cloud removed versus time for various rain rates.

result may develop varying nucleation characteristics with time. Also, the soluble fission products that attach to the debris particle surface during particle formation may influence the nucleation characteristics.

In the latter stages of fireball development a visible cloud of water or ice usually forms. Any debris particles that participate in this formation of water droplets or ice particles probably will be good cloud condensation nuclei if the induced natural cloud evaporates and the particle is then entrained into a rain cloud environment.

Cloud Models and Rainout

The life history of radioactive debris injected into the atmosphere is greatly altered if the debris enters a region of precipitation: the amount of debris deposited on the ground can be greatly enhanced and the vertical distribution is completely changed. The two processes associated with clouds that are primarily responsible for determining the amount and distribution of debris deposited on the ground are (1) the microphysical process by which airborne debris is collected by drops, and (2) the dynamic process that transports debris into the natural cloud, thus allowing the microphysical interactions to occur. These two processes cannot be separated, since the integrated effects of the microphysics of raindrops, i.e., evaporation, condensation, drop collisions, melting, and freezing, produce the essential driving forces for the dynamic situation that tends toward supersaturation and the existence of droplets in unsaturated regions. Therefore, it is necessary to be able to describe the combined action of both microphysical and dynamic processes in order to evaluate the effects of precipitation on the scavenging of radioactive debris particles. By the use of numerical cloud models with an appropriate parameterization of the microphysics, it is possible to describe the life cycle of convective cells, which produce the great majority of the world's rainfall.

One of the characteristics of clouds that differentiates them from their environment is the existence of enhanced vertical motions. The noncloudy atmosphere seldom has vertical velocities greater than a few centimeters per second; a typical convective cell has upward motions of several meters per second. Convective cells are frequently observed to develop upward from a small cumulus cloud, and they have rather sharp side boundaries indicating that there is little transport across their sides. This characteristic of convective clouds has led to the formulation of a one-dimensional entrainment model of a convection cloud in which air is assumed to rise vertically from cloud base with only a small amount of mixing (entrainment) across its sides. The one-dimensional model cloud is assumed to be made up of a stack of cylindrical slabs, all of the same thickness ΔZ. The average values of the relevant variables, e.g., liquid water content, temperature, velocity, radius, etc., are evaluated from the corresponding equations of motion, energy, and continuity for each of these slabs, giving a vertical profile for each variable. The equations can be integrated in time to provide a time-history of the formation, growth, and decay of a convective cell. The microphysical processes are represented in such a model by terms in the continuity equations that describe the production or destruction of the appropriate phase of water substance because of each microphysical interaction. The presence of radioactive debris has been included in this model by adding appropriate equations of continuity with the proper representation of microphysical interactions between debris and drops. In such a model, debris is drawn in through the base of the cloud and a small amount of debris is entrained across the horizontal boundaries. The debris that enters the cloud is subjected to the various microphysical scavenging mechanisms, so that some of the radioactive particles are collected by the rain and brought to the ground.

The enhanced vertical transport of air inside clouds makes it possible for the vertical distribution of debris inside the cloud to differ greatly from that outside the cloud. Since the air inside the cloud originated primarily at levels near the cloud base, its debris concentration tends to be characteristic of this level. If the debris is initially distributed in such a way that the concentration is highest in the vicinity of the cloud base, the total amount of debris inside the cloud would be several times as large as that in a similar volume of air outside this cloud. In this situation, the potential for high levels of radioactivity on the ground as a result of precipitation scavenging is enhanced by the field of motion of the cloud. Of course, if the debris is above the cloud top it is not subject to precipitation scavenging. The model was run several times with the initial distribution of debris uniform in the horizontal and Gaussian in the vertical, which has peak concentration at height H, and a standard deviation of 560 m. The most relevant ratios (Table 1) are those at 25 and 50 min when rain is falling and scavenging is most efficient; they show that when the debris is initially at low levels, the potential for scavenging is enhanced, but when the

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The rain cloud is assumed to be much smaller in horizontal extent than the debris cloud at the time of interaction.

Table 1. Ratio of debris inside cloud to debris outside cloud in similar volumes.

Initial debris height (m)	Time (min)			
	0	25	50	75
1340	1.00	1.76	1.83	1.51
1740	1.00	1.36	1.48	1.34
2140	1.00	0.66	0.75	0.97
2440	1.00	0.31	0.36	0.69
2840	1.00	0.20	0.21	0.50

debris is initially well above cloud base (1400 m) the potential for scavenging is greatly reduced. In addition, current knowledge indicates that the most important link in the scavenging process is consensation nucleation, and for convective clouds this process occurs efficiently only in the region of the cloud base.

Work is proceeding on modeling the scavenging mechanism so that the accumulation of debris on the ground is calculated. Preliminary results indicate (1) that the scavenging is efficient for low-level debris, and (2) that the total amount of debris deposited by a single convective cell on a unit area of ground can be several times as large as the amount initially in a vertical column of unit cross-sectional area directly overhead. That is, the cloud can act as a mechanism for concentrating debris.

The one-dimensional model has provided considerable insight and has indicated several important aspects of scavenging, but it is a crude model that does not adequately describe the dynamic state of the atmosphere. Consequently, effort is being expended to develop two-dimensional models of convection with scavenging, in order to more completely evaluate the interaction between radioactive particles and rain clouds. The one-dimensional model cannot adequately describe the convergence of air at low levels and divergence at high levels that is a characteristic of convection; a two-dimensional axisymmetric model is being modified to include scavenging so that this part of the process can be evaluated more correctly. Another very important feature of the atmosphere that affects precipitation scavenging is changes of wind speed (and direction) with height, i. e., wind shear. Therefore, a two-dimensional rectangular model is being modified to include scavenging of radioactivity by a cloud growing in an environment with speed shear due to the horizontal wind. In spite of its shortcomings, however, the one-dimensional model does provide an adequate and convenient framework in which to evaluate the effects of various parameterizations of the scavenging process, and we will continue to use it for that purpose.

Controlling the Process

If the rainout mechanism is correctly described above, then it may be possible to substantially reduce the rainout coefficient (and thus reduce the dose received at any given point) by rendering the debris particles nonwettable. This would be done by coating the particles with a water repellent surfactant as has been described in UCRL-51328.[2] Experimental verification of the mechanism is desirable.

The solid particles formed in a nuclear detonation are metal oxides in the form of crystals or glasses and possibly some free metallic particles. All such particles have large surface energies compared to that of water (73 ergs/cm^2) (see Table 2). Thus, one would expect water to wet and spread on them[8-10] and from the Fletcher analysis[3] all of them would be active nucleating agents in a cloud if they

Table 2. Surface energies of various materials, taken from references cited.

Materials	γ at 0 K (erg cm^2)	Reference
Oxides of general formula MO		
MgO	1090	13
FeO	1060	13
MnO	1010	13
CaO	820	13
SrO	700	13
BaO	604	13
BeO	> 1420	13
CdO	530	13
ZnO	600	13
PbO	250	13
General formula M_2O_3		
B_2O_3	79.5 @ 900°C	14
Al_2O_3	905 @ 1850 C	15
General formula MO_2		
UO_2	642 ± 20%	13
ZrO_2	800 ± 20%	13
ThO_2	530 ± 20%	13
SiO_2	> 800	13
TiO_2	> 800	13

were 0.1 μm in radius or larger. Their rates of nucleation can be reduced by the addition of low-surface-energy materials that will adsorb onto the particle surfaces and lower their surface energies to less than that of water. A monolayer of perfluoro fatty acid on platinum reduces its specific surface free energy[11,12] from about 2 000 ergs/cm^2 to about 10 ergs/cm^2; water should not wet or spread on particles so treated. Even a small departure from perfect wettability should hinder the nucleation dynamics.

Coating nuclear debris in the atmosphere would involve major problems in the delivery of the surfactant and in mixing it with the debris cloud. These problems cannot be addressed until the required concentrations of the surfactant and its physical characteristics are specified. This section, therefore, deals only with the selection and testing of possible coating materials.

Any coating materials designed for atmospheric use either must be stable in the photochemical environment of the atmosphere (for at least a few hours) or must yield products that are themselves good coating materials. This condition may serve to eliminate many organic compounds.

Also, the coating material must be sufficiently volatile so that it can be added to the cloud as a vapor (so that the mixing will not be determined by the relatively slow particle diffusion process). This eliminates many nonvolatile oils and suggests that stable organic compounds of moderate molecular weight (i.e., 100 to 400) will be likely to provide the most reasonable choices.

A selection of compounds with vapor pressure ≥ 1 × 10^{-2} Pa (10^{-4} Torr) at ~280 K and with reasonable photochemical stability could be made on the basis of (1) their effectiveness in rendering bulk surfaces nonwettable, (2) measurements of the effect of small quantities of these species on the rate of growth of synthetic laboratory aerosols; and (3) their photochemical stability as coatings, measured on bulk surfaces and laboratory aerosols. Experiments on laboratory aerosols would be used also to establish the concentration of the surfactant necessary to produce the desired effect.

As was shown by Fulk (in Ref. 2), this concentration can be quite low. If the heat of adsorption is high, the actual mass M of material needed to provide a complete monolayer on the particles from a device is

$$M \cong C_s \cdot \frac{W_0}{\rho} \cdot \left(\overline{\frac{1}{r}}\right) \cdot mw/N_0,$$

where C_s is the surface concentration required in molecules per square centimeter, W_0 is the mass of the device converted to oxides, ρ is the density of debris particles, $\left(\overline{\frac{1}{r}}\right)$ is the mean reciprocal radius, mw is the molecular weight of the additive, and N_0 is Avogadro's number. If

$$C_s = 10^{15}, \quad W_0 = 10^6 g, \quad \rho = 3.5,$$

$$\left(\overline{\frac{1}{r}}\right) = 2 \times 10^5, \text{ and } mw \cong 300,$$

then

$$M \cong 10^{15} \times \frac{10^6}{3.5} \cdot 2 \times 10^5$$

$$\cdot\ 300/(6 \times 10^{23}) \cong 2.8 \times 10^4 \text{ g or } 28 \text{ kg.}$$

The value of C_s used here is an upper limit; all of the other numbers are reasonable estimates, but conservative in the sense that they are chosen to give high rather than low values for M.

Let us consider S_0 the quantity of coating material necessary to provide a concentration that, if well mixed with the debris cloud, would result in deposition of the full monolayer within a reasonable period of time. We then have the requirement that

$$C_s \leq \int_{t_0}^{t_{max}} J_s \, dt,$$

where t_{max} is the maximum permissible time, t_0 is the injection time and J_s is the flux of additive per unit of aerosol surface area. This is approximately given by

$$C_s \leq \int_{t_0}^{t_M} \gamma \, C(t) \, \overline{S} \, dt,$$

where γ is the sticking coefficient (assumed equal to unity), C(t) is the concentration as a function of time, and $\overline{S}$ is the mean thermal speed of surfactant molecules ($\sim 10^4$ cm/sec) · C(t) will be approximately

$$\frac{(S_0 - A(t))}{V(t)},$$

where V(t) is the volume of the debris cloud and A(t) is the flux to the surface prior to time t. If A(t) is small relative to S_0, this consideration of fluxes is not an important additional constraint, thus A(t) will be omitted. For a 10-kt device,

$$V(t) \simeq 3.2 \times 10^{16} e^{+t/3300} \text{ cm}^3$$

so that we have

$$10^{15} \leq \int_{t_0}^{t_{max}} \frac{S_0 \times 10^4}{3.2 \times 10^{16} e^{+t/3300}} \cdot 4.$$

For t_0 = 300 sec and t_{max} = 3000 s, S_0 must be approximately 10^{25} molecules. Thus, this constraint is a trivial addition to the quantity needed to coat the particles.

If the heat of adsorption is not of the order of 20 kcal/mole or larger, desorption may be an important process, and it becomes necessary to calculate the equilibrium concentration at the surface as the product of the flux (as described above) and the "sitting time" τ.

$$C_s = J_s \times \tau$$

where τ is given by

$$\tau = \tau_0 \, e^{\Delta H/RT}$$

$$\approx 10^{-13} \, e^{\Delta H/RT}$$

where ΔH is the heat of adsorption, R is the gas constant, and T the temperature.

This suggests that materials that have low vapor pressures or that are chemisorbed on oxides should be given the most consideration. If the quantity necessary to coat the natural aerosol burden at a height of ~2 km over regions of potential rainout concern is estimated, it would appear that such considerations could increase the amount of necessary injection by a factor of at most 2 to 10. Thus if a suitable material were to be determined and the problems of delivery and mixing are not insurmountable, not more than about 500 kg of material would be required for coating, and perhaps about 50 kg would be adequate.

There are several other possible effects of additives on cloud dynamics about which little or nothing is known. Research into these effects is needed. It should be kept in mind also that since a cloud is a dynamic system, the applicability of equilibrium arguments is somewhat questionable and may at times be misleading.

It is probable that the condensation and evaporation coefficients of water molecules at surfaces of liquid water and/or ice can be modified by as much as a factor of 10 by contaminating the surfaces.[16-18] Many different combinations of factors are involved in the interaction between incident water molecules and surfaces of water drops and ice particles:

- Specific condensation coefficient.
- Specific evaporation coefficient.
- Thermal accommodation coefficient.
- Momentum accommodation coefficient.
- Extent of compliance with the cosine law for diffuse reflection.
- Extent the cosine law would obtain due to scattering from surface irregularities alone.

Neither the molecular and/or atomic interaction mechanisms of wetting nor the wetting rates are well understood. Electrification makes the problem even more complex. A number of efforts are needed to understand the off-equilibrium dynamics of clouds. Among the simpler experiments which might be attempted are determinations of the effects of additives on aerosol nucleation and growth rates.

Conclusions

Much work remains to be done to make the definitive assessment of the potential hazards from rainout. It is recognized that not all agencies agree on the efficiency of the scavenging process and the removal rate of debris particles. In particular, the particle size distribution for nuclear clouds in the yield range of interest deserves an improved experimental basis. It is expected that work over the next six months will provide a more precise basis for analysis of the phenomenon.

Acknowledgments

This report is the product of the efforts of a research team at Lawrence Livermore Laboratory currently working on an assessment of the potential hazards of precipitation scavenging from nuclear debris clouds. In addition to the authors, the following persons made major contributions to this paper: C. R. Molenkamp, K. R. Peterson, E. V. Jankus, W. H. Duewer, M. M. Fulk, and R. C. Orphan. The work was performed in part under the auspices of the U. S. Atomic Energy Commission and in part under the sponsorship of the Defense Nuclear Agency (Contract IACRO-73-814).

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